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Longitudinal spin fluctuations in itinerant electron ferromagnets

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Abstract. The relaxational dynamics of weak itinerant magnets below the Curie temperature is investigated. The calculation is based on the non-linear equations of motion for the magnetic order parameter. It is shown that besides the conventional linear relaxation mechanism due to the Landau damping of spin fluctuations (SFs) which gives rise to overdamped paramagnons, there may exist a different mechanism which is due to non-linear three-mode interactions of spin fluctuations. The interplay between these two mechanisms is analysed and it is argued that three-mode scattering with emission, or absorption, of a longitudinal SF by a magnon may dominate. This results in a new type of overdamped longitudinal SF propagating close to the magnon dispersion, which has probably been observed in recent neutron scattering experiments.

1. Introduction

The last decade has contributed much to our understanding of the spin fluctuation (SF) phenomena in weak itinerant ferromagnets close to a magnetic instability. The thermodynamics accounting for both types of SF, weakly damped magnons [1] and overdamped paramagnons [2, 3] arising in the Stoner continuum below and above the Curie temperature, was worked out based on the weak-SF-coupling approximation. Recently the SF thermodynamics of itinerant magnets was generalized to account for the strong spin anharmonicity effects caused by the large zero-point SF amplitudes [4, 5].

However, up to now the investigation of the SF dynamics of itinerant magnets outside the critical region has mainly been restricted to a non-dissipative (spin wave) regime described by the non-linear Landau–Lifshitz [6] and dynamical magnon equations which were derived within the Fermi liquid concept [1, 7]. The dynamics of the overdamped SFs, paramagnons, was actually described on the basis of the linear, or random phase approximation, which in the long-wavelength low-frequency limit results in the following form for the transverse $\chi_t(k)$ and longitudinal $\chi_l(k)$ dynamical susceptibilities [2, 3],

$$\chi_\nu^{-1}(k) = \chi_\nu^{-1} + ck^2 - \mathrm{i}\frac{\omega}{\Gamma_0(\boldsymbol{k})} \qquad (\nu = \mathrm{t, l}) \tag{1}$$

where $k = (\omega, \boldsymbol{k})$, ω and $\boldsymbol{k}$ are the frequency and wavevector of the SF and $\chi_{t,l}$ are the magnetization M and temperature T dependent susceptibilities. Here c and $\Gamma_0(\boldsymbol{k})$ account for the spatial dispersion and relaxation rate, respectively. In the collisionless Fermi liquid regime the relaxation rate was assumed to be linear in $\boldsymbol{k}$, $\Gamma_0(\boldsymbol{k}) = \Gamma_0|\boldsymbol{k}|$ with the coefficient Γ_0 related to the Landau damping of SF, which in weak itinerant magnets is independent of M, T, ω and $\boldsymbol{k}$ [2, 3].

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On the other hand, the non-linear coupling of SFs which essentially influences thermodynamics of itinerant magnets [1–5] may give rise to multimode scattering processes and affect the scattering rate $\Gamma_0(k)$ of SFs, as can be seen from investigations of critical dynamics (see, e.g., [8]). Recently we have shown that three-mode scattering processes with the absorption or emission of a longitudinal SF by a magnon dominate the magnon liftime in weak itinerant ferromagnets [9]. Similar processes were also shown to affect spectral properties of longitudinal SFs in Heisenberg ferromagnets [10]. In this paper we analyse non-linear three-mode interactions of SFs with respect to the spectrum of longitudinal SFs of itinerant magnets below the Curie temperature.

2. Non-linear magnetic dynamics

We describe the magnetic dynamics of an isotropic weak itinerant ferromagnet by the following non-linear equations of motion for the spatially dependent and Fourier transformed magnetic order parameter,

$$\chi_l^{-1}(k)m_l(k) = -\sum_{k_1+k_2=k}[\tfrac{1}{2}W_{ltt}(-k,\,k_1,\,k_2)m_t(k_1)m_t^*(-k_2) + W_{lll}(-k,\,k_1,\,k_2)m_l(k_1)m_l(k_2)] + \ldots \tag{2}$$

$$\chi_t^{-1}(k)m_t(k) = -\sum_{k_1+k_2=k}W_{ltt}(k_1,\,k_2,\,-k)m_l(k_1)m_t(k_2) + \ldots \tag{3}$$

where we retain only terms which are bilinear in the longitudinal $m_l(k)$ and transverse $m_t(k)$ SF amplitudes. Here

$$\sum_{k} = \sum_{k}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}$$

W_{ltt} and W_{lll} characterize the amplitudes of three-mode scattering processes.

The dynamical susceptibilities $\chi_{t,l}(k)$ in the paragmagnon region are given by equation (1) provided the wavevector of transverse SF is above the maximum magnon wavevector k_0. It separates the spin wave and paramagnon regions and in weak itinerant magnets is approximately defined by the intersection of the magnon dispersion with the Stoner continuum, $k_0 \approx (p_+ - p_-)/\hbar$, where p_σ is the Fermi momentum of electrons with spin $\sigma = \pm$ (see, e.g., [1–3]) In the spin wave region, $|k| < k_0$, we have

$$\chi_t(k) = \frac{2\mu_B M}{\hbar\left[\omega_m(k) - \omega - i\tau^{-1}(k)\right]} \tag{4}$$

where $\omega_m(k) = Dk^2$ and $\tau(k)$ are the frequency and lifetime of magnons, $D = (2\mu_B M/\hbar)c$ is the stiffness constant, and μ_B is the Bohr magneton. As was reported recently [9], the magnon lifetime $\tau(k)$ in weak itinerant magnets is dominated by processes of scattering by longitudinal SFs, and exept for low temperatures has a $\sim k^2\ln(1/k^2)$ wavevector dependence. Not very close to T_c, outside the critical region, the magnon damping is relatively weak, and magnons may be regarded as a propagating mode, whereas the spin wave region of the Brillouin zone vanishes as $T \to T_c$. Formulae (2) and (3) were derived from the non-linear equations of motion of a Fermi liquid by integrating out the individual quasiparticle variables

[1, 7]. They were also obtained directly from the Ginzburg–Landau effective Hamiltonian [9]. In the low-frequency long-wavelength limit ($\omega, \boldsymbol{k} \to 0$, $\omega/|\boldsymbol{k}| \to 0$) and in the lowest order in M both approaches yield the same results for the scattering amplitudes,

$$W_{\rm ltt} = W_{\rm ttl} = 2\gamma M$$

$$W_{\rm lll} = 3\gamma M \tag{5}$$

where γ is the SF coupling constant which defines the $\gamma M^4/4$ contribution to the Landau free energy. The non-linear terms in the RHS of (2) and (3) may contribute essentially to the magnetic relaxation. The renormalized relaxation rate, $\Gamma_\nu(k, T)$, which should replace $\Gamma_0(\boldsymbol{k})$ in (1), depends, apart from the wavevector, on the frequency and polarization of the SF and is given by

$$\Gamma_\nu^{-1}(k, T) = \Gamma_0^{-1}(\boldsymbol{k}) + \Gamma_{\rm n\nu}^{-1}(k, T) \tag{6}$$

where $\Gamma_{\rm n\nu}(k, T)$ defines the contribution arising from the non-linear interactions of SFs. In the lowest-order approximation accounting for three-mode interactions we have

$$\Gamma_{\rm nl}^{-1}(k, T) = \frac{2\hbar}{\omega} \sum_{\nu=t,l} \sum_{k'} |W_{\rm l\nu\nu}(-k, k+k', -k')|^2 {\rm Im}\,\chi_\nu(k'){\rm Im}\,\chi_\nu(k+k')(N_{\omega'} - N_{\omega+\omega'}) \tag{7}$$

$$\Gamma_{\rm nt}^{-1}(k, T) = \frac{2\hbar}{\omega} \sum_{k'} |W_{\rm ltt}(-k, k+k', -k')|^2 {\rm Im}\,\chi_{\rm t}(k'){\rm Im}\,\chi_{\rm l}(k+k')(N_{\omega'} - N_{\omega+\omega'}) \tag{8}$$

where $N_\omega = [\exp(\hbar\omega/k_{\rm B}T) - 1]^{-1}$. (7) describes damping of a longitudinal SF due to the processes of absorption (emission) by a transverse ($\nu = $ t) or longitudinal ($\nu = $ l) SF, and equation (8) accounts for damping of a transverse SF caused by the absorption (emission) of a longitudinal SF. It should be mentioned that these three-mode scattering processes are analogous to those discussed in the critical dynamics [8] and also in the Heisenberg ferromagnets outside the critical region [10].

Here we focus on the most important contribution to $\Gamma_{\rm nl}(k, T)$ (below the Curie temperature $T_{\rm c}$) arising from the emission (absorption) of a longitudinal SF by a magnon. After substituting (4) and (5) into the expression (7) the integration over ω' and $\boldsymbol{k}'$ is easily performed, provided magnons are weakly damped, $\omega_{\rm m}(\boldsymbol{k})\tau(\boldsymbol{k}) \gg 1$. This yields the following non-linear contribution to the relaxation rate of the longitudinal SF,

$$\Gamma_{\rm nl}(k, T) = \Gamma_{\rm n}|\boldsymbol{k}|L^{-1}(k, T)\hbar\omega/k_{\rm B}T \tag{9}$$

where $\Gamma_{\rm n}$ and $L(k, T)$ are given by

$$\Gamma_{\rm n} = \frac{16\pi}{\hbar}(cM\chi_{\rm l})^2 \tag{10}$$

$$L(k, T) = \ln\left[\frac{\sinh\left\{\hbar\left([\omega + \omega_{\rm m}(\boldsymbol{k})]^2 + \tau^{-2}(\boldsymbol{k})\right)/8k_{\rm B}T\omega_{\rm m}(\boldsymbol{k})\right\}}{\sinh\left\{\hbar\left([\omega - \omega_{\rm m}(\boldsymbol{k})]^2 + \tau^{-2}(\boldsymbol{k})\right)/8k_{\rm B}T\omega_{\rm m}(\boldsymbol{k})\right\}} \times \frac{\sinh\{\hbar\omega_{\rm max}/2k_{\rm B}T\}}{\sinh\{\hbar(\omega + \omega_{\rm max})/2k_{\rm B}T\}}\right] \tag{11}$$

and $\omega_{\max} = \omega_m(k_0)$ is the maximum magnon frequency. We emphasize that the non-linear contribution to $\mathrm{Im}\,\chi_l^{-1}(k)$ given by formulae (9) – (11) does not depend on the form of the longitudinal SF spectrum and incorporates only spin wave parameters. This result is similar to that reported previously for the longitudinal correlation function of a Heisenberg ferromagnet [10]. Analogously to the correlation function of [10] the inverse relaxation rate, $\Gamma_{nl}^{-1}(k, T)$, in the absence of magnon damping has a logarithmic divergence near the magnon frequencies $\omega = \pm\omega_m(k)$ and in the long-wavelength limit increases $\sim 1/|k|$. Vanishing of $\Gamma_{nl}(k, T)$ in this limit reflects the conservation of the total magnetization in isotropic magnets, which is not affected by the scattering processes considered here.

It should be mentioned that in the long-wavelength limit both contributions to the relaxation rate, linear and non-linear ones, have similar linear dependences on the wavevector. In the collisionless regime this k dependence of $\Gamma_0(k)$ arises due to Landau damping when SFs are absorbed by Fermi quasiparticles. Analogously, the linear k dependence of $\Gamma_{nl}(k)$ is caused by the absorption of SFs by weakly damped magnons. It is worth to noting that these relaxation mechanisms are usually neglected in the analysis of critical dynamics. For example, in the conventional van Hove theory the relaxation rate is taken in the form proportional to the squared wavevector [8], which in itinerant magnets may be related to the relaxation mechanism due to scattering of electrons by impurities [2]. In this paper we restrict ourselves to the collisionless Fermi liquid regime in weak itinerant magnets, far from the critical region.

3. Spectrum of longitudinal spin fluctuations

Substituting (6) and (9) instead of $\Gamma_0(k)$ into (1) yields the following expression for the imaginary part of the longitudinal dynamical susceptibility,

$$\mathrm{Im}\chi_l(k) = \chi_l(k)\frac{\omega_{SF}(k)[\omega + \omega_{st}(k, T)]}{\omega_{SF}^2(k) + [\omega + \omega_{st}(k, T)]^2} \qquad (12)$$

which defines the longitudinal SF spectrum and the intensity of inelastic neutron scattering (see, e.g., [11–15]). Here $\chi_l(k) = \left(\chi_l^{-1} + ck^2\right)^{-1}$ is the static susceptibility,

$$\omega_{SF}(k) = \Gamma_0|k|\chi_l^{-1}(k) \qquad (13)$$

is the conventional characteristic frequency of paramagnons [2, 3], and the frequency

$$\omega_{st}(k, T) = \omega\frac{\Gamma_0(k)}{\Gamma_{nl}(k, T)} = \frac{k_B T}{\hbar}\frac{\Gamma_0}{\Gamma_n}L(k, T) \qquad (14)$$

describes the effects of the non-linear three-mode scattering of longitudinal SFs by magnons. Formula (13) accounts for both relaxation mechanisms, a linear one due to Landau damping and a non-linear one caused by three-mode interactions of SFs. Below we analyse the interplay between them, e.g., for $\omega > 0$.

According to (11) and (14) the frequency $\omega_{st}(k, T)$ is a non-monotonic function of ω. In the low-frequency limit $\omega \to 0$ it increases linearly with ω, diverges logarithmically at $\omega = \omega_m(k)$ and is small for $\hbar\omega \gg k_B T$. The effects of magnon damping result in a finite maximum of ω_{st} at $\omega = \omega_m(k)$. The non-monotonic dependence of $\omega_{st}(k, T)$ on the frequency ω may essentially change the shape of the longitudinal SF spectrum with respect to the simple Lorentzian which we get omitting three-mode interactions of SFs [2, 3].

According to (11)–(14) $\mathrm{Im}\,\chi_1(k)$ may have several maxima at frequencies ω given by the equation

$$\omega + \omega_{st}(k, T) = \omega_{SF}(\boldsymbol{k}). \tag{15}$$

Besides that $\mathrm{Im}\,\chi_1(k)$ has two extrema when ω satisfies the condition

$$\partial\,[\omega + \omega_{st}(k, T)]\,/\partial\omega = 0. \tag{16}$$

Below we briefly analyse the shape of the longitudinal SF spectrum for relatively long wavelengths, when

$$\omega_{SF}(\boldsymbol{k}) \gg \omega_{m}(\boldsymbol{k}). \tag{17}$$

A detailed analysis of the spectral properties of SFs with account of non-linear interactions will be published elsewhere. According to (12) the shape of the spectrum is crucially dependent on the parameter

$$\eta_1(\boldsymbol{k}) = \{\omega_m(\boldsymbol{k}) + \omega_{st}[\omega = \omega_m(\boldsymbol{k})]\}/\omega_{SF}(\boldsymbol{k}). \tag{18}$$

At relatively large wavevectors, when

$$\eta_1(\boldsymbol{k}) \leqslant 1 \tag{19}$$

equation (15) has only one solution approximately given by

$$\omega = \omega_{SF}(\boldsymbol{k}). \tag{20}$$

At this frequency the SF spectrum (12) has a broad maximum with

$$\mathrm{Im}\,\chi_1(k) = \tfrac{1}{2}\chi_1(\boldsymbol{k}) \tag{21}$$

which corresponds to conventional paramagnon excitations. According to (16), $\mathrm{Im}\,\chi_1(k)$ has also a rather sharp maximum near

$$\omega = \omega_m(\boldsymbol{k}) \tag{22}$$

caused by non-linear interactions with magnons. The height of the maximum is strongly dependent on the ratio

$$\eta_2(\boldsymbol{k}) = \omega_{st}\,[\omega = \omega_m(\boldsymbol{k})]\,/\omega_m(k) \tag{23}$$

and is less than the value given by equation (21).

At longer wavelengths, when

$$\eta_1(\boldsymbol{k}) > 1 \tag{24}$$

equation (15) has three solutions: two of them are shifted somewhat above and below the frequency $\omega = \omega_m(k)$; the third one is close to the paramagnon frequency $\omega = \omega_{SF}(\boldsymbol{k})$. At these frequencies $\mathrm{Im}\,\chi_1(k)$ has maxima with heights given by equation (21). So, with the increase of $\eta_1(k)$ the maximum at $\omega \approx \omega_m(\boldsymbol{k})$ caused by non-linear SF interactions is

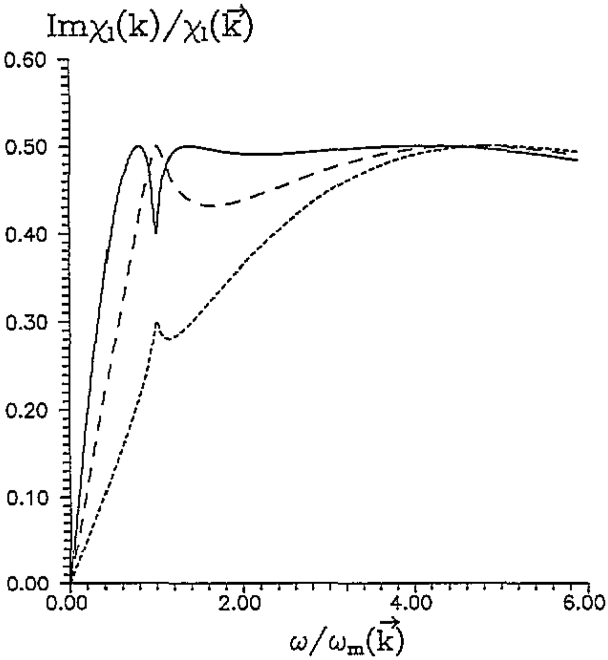

Figure 1. Spectrum of longitudinal spin fluctuations calculated from (12) with $\omega_{max}/\omega_m(k) = \omega_m(k)\tau(k) = k_BT/\hbar\omega_m(k) = 100$ and $\omega_{SF}(k)/\omega_m(k) = 5$ for $\eta_1(k) = \frac{1}{3}$ and $\Gamma_0/\Gamma_n = 7.25 \times 10^{-4}$ (- - -); $\eta_1(k) = 1$ and $\Gamma_0/\Gamma_n = 4.35 \times 10^{-3}$ (– – –); $\eta_1(k) = 2$ and $\Gamma_0/\Gamma_n = 9.78 \times 10^{-3}$ (——).

split into two, below and above $\omega_m(k)$, being accompanied by a minimum at $\omega \approx \omega_m(k)$. Figure 1 illustrates the spectrum of longitudinal SFs and the interplay between the two relaxation mechanisms related to the linear Landau damping and non-linear interactions of SFs, respectively.

The fine structure of the longitudinal SF spectrum near the magnon frequencies $\omega_m(k)$ becomes very transparent for long wavelengths and (or) in the limit $\Gamma_0/\Gamma_n \to \infty$, when

$$\eta_2(k) \gg 1. \tag{25}$$

In this limit the SF spectrum is dominated by the non-linear relaxation mechanism, and Landau damping plays a negligibly small role. Neglecting the effects of Landau damping in (12) we arrive at the following expression for the longitudinal SF spectrum near $\omega = \omega_m(k)$,

$$\mathrm{Im}\,\chi_l(k) = \chi_l(k)\frac{\omega_n(k)\omega_T(k, T)}{\omega_n^2(k) + \omega_T^2(k, T)} \tag{26}$$

where the frequencies

$$\omega_n(k) = \Gamma_n|k|\chi_l^{-1}(k) \qquad \omega_T(k, T) = \frac{k_BT}{\hbar}L(k, T) \tag{27}$$

characterize the non-linear relaxation. We emphasize that the SF spectrum defined by (26) may be interpreted in terms of a new type of overdamped collective spin excitation with frequencies $\omega \approx \omega_m(k)$. These excitations are related to the three-mode non-linear SF interactions and result from emission (absorption) of a longitudinal SF by a magnon. The regime of the non-linear magnetic relaxation is illustrated by figure 2.

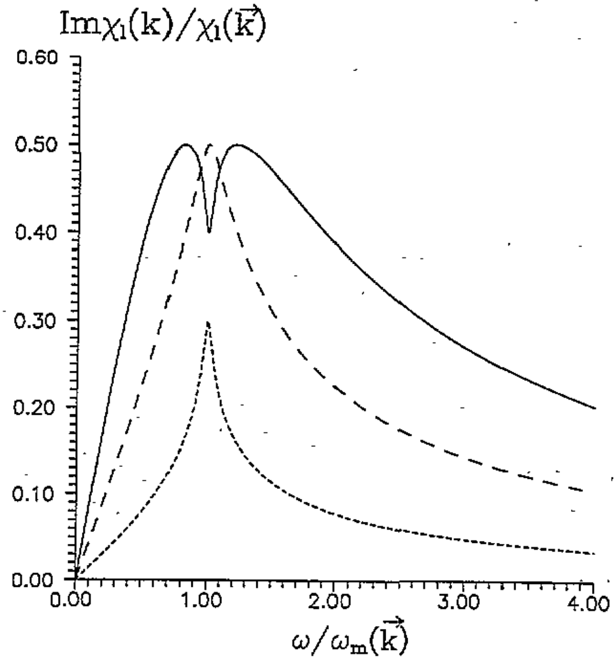

Figure 2. Spectrum of longitudinal spin fluctuations calculated from (12) with $\omega_{max}/\omega_m(k) = \omega_m(k)\tau(k) = k_B T/\hbar\omega_m(k) = 100$ and $\Gamma_0/\Gamma_n = 0.1$ for $\eta_l(k) = \frac{1}{3}$ (- - -); $\eta_l(k) = 1$ (— — —); $\eta_l(k) = 2$ (——).

4. Discussion and summary

Using the recent neutron scattering data for itinerant magnets [3, 11–15] it is possible to estimate the role of the non-linear three-mode interactions of SFs and to find some evidence for the new type of longitudinal SF with frequencies close to magnon frequencies $\omega = \omega_m(\boldsymbol{k})$, as discussed above. With the neutron scattering and magnetic measurement data from [3] [11] and [12] we estimate Γ_0/Γ_n to be 0.4, 1×10^{-2} and 8×10^{-3} for the weak itinerant magnets MnSi, Ni_3Al and $ZrZn_2$, respectively. This suggests that the main relaxation mechanism in the major part of the Brillouin zone for these materials is due to Landau damping. Corresponding excitations are conventional paramagnons which is confirmed by neutron scattering experiments [3, 12].

The non-linear relaxation mechanism manifests itself when Landau damping of SFs is negligible, or at rather long wavelength, e.g. for $\eta_2(\boldsymbol{k}) > 1$, when

$$k \leqslant \sqrt{\frac{\Gamma_0}{\Gamma_n}\frac{k_B T}{\hbar D}} \tag{28}$$

where D is the magnon stiffness constant defined above.

For the weak ferromagnets mentioned above the inequality (28) is satisfied for a rather long-wavelength region which up to now has not been investigated by neutron scattering experiments. For example, for $ZrZn_2$ with [12] $D = 90$ meV Å^{-2} at $T \simeq T_c/2$ we have from equation (28) $k \leqslant 1 \times 10^{-2}$ Å^{-1}. Nevertheless, appreciable damping of spin waves measured at shorter wavelengths may be attributed to the non-linear three-mode SF interactions as well as to the conventional Landau damping mechanism giving rise to overdamped longitudinal SFs near magnon frequencies as discussed above.

On the other hand, recent inelastic polarized neutron scattering experiments in amorphous $Fe_{86}B_{14}$ and $Fe_{40}Ni_{40}P_{14}B_6$ itinerant magnets revealed overdamped longitudinal SFs with frequencies close to the magnon frequencies (22) and wavevectors $k \sim 0.1$ Å^{-1} [14]. These excitations were observed both near and far below the Curie temperature with the ratio of spin wave to longitudinal SF scattering intensities being 2.5:1. The reported data [13] for the crystalline Invar system $Fe_{65}Ni_{35}$ also suggests the existence of overdamped longitudinal SFs propagating close to the magnon frequencies though they were not clearly seen in more recent measurements [15].

In our view, recent neutron scattering experiments [14] present some evidence for the new type of longitudinal SF with frequencies close to the magnon frequency $\omega_m(\boldsymbol{k})$, discussed in this paper, at least in the case of amorphous itinerant magnets. Inelastic neutron scattering experiments [13, 15] also revealed the fact that the measured data for these materials could not be explained on the basis of conventional paramagnon excitations. This also supports the existence of a relaxation mechanism different from the conventional Landau damping mechanism, which—on the other hand—dominates the magnetic dynamics of weak itinerant ferromagnets of the $ZrZn_2$ type.

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